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A SURFACE PLASMON DISPERSION RELATION IN THE PRESENCE OF SPATIAL DISPERSION*

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ABSTRACT

A dispersion relation for surface plasmons (retardation neglected) is obtained in the presence of spatial dispersion. The physical system considered is one of vacuum separated by a plane interface from a dielectric medium occupying the half-space $x_3 > 0$, and characterized by a nonlocal dielectric function $\epsilon(\vec{k}_\parallel\omega|x_3x_3')$, assumed to be symmetric in x_3 and x_3'. Here $\vec{k}_\parallel$ is a two-dimensional wave vector whose components are parallel to the interface, and ω is the frequency of the electromagnetic field in the medium. The dispersion relation has the form $1 + k_\parallel\chi(\vec{k}_\parallel\omega|x_3 = 0,\ x_3' = 0) = 0$, and an explicit prescription for obtaining the function $\chi(\vec{k}_\parallel\omega|x_3x_3')$ is presented. The use of the dispersion relation is illustrated by applying it to two examples: (1) a local dielectric constant; and (2) the nonlocal dielectric function used previously by Maradudin and Mills. In both cases previously obtained results are recovered.

I. Introduction

Several years ago Ritchie and Marusak[1] published a derivation of a dispersion relation for surface plasmons[2] in the presence of spatial dispersion. Their result has the simple form

$$1 + \frac{k_\parallel}{\pi} \int_{-\infty}^{\infty} \frac{dk_3}{k^2 \epsilon(\vec{k},\omega)} = 0 \quad . \tag{1.1}$$

Here $\epsilon(\vec{k},\omega)$ is the frequency and wave vector dependent dielectric constant of the material out of which the semi-infinite medium is composed, along whose planar interface with vacuum the surface plasmon propagates. In Eq. (1.1) $k_\parallel$ is the magnitude of the projection of the three-dimensional wave vector $\vec{k}$ on the plane of the surface, and k_3 is the component of $\vec{k}$ normal to the surface.

A central assumption in the derivation of Eq. (1.1) is that the electrons in the semi-infinite dielectric medium are reflected specularly from the surface. It is this assumption which ultimately leads to the appearance of the bulk dielectric constant $\epsilon(\vec{k},\omega)$ in Eq. (1.1).

However, in any real solid it is unlikely that all of the electrons are reflected specularly from the surface. It is more likely that some fraction are scattered specularly and the rest diffusely, or that more general boundary conditions obtain. In any case, it would seem to be desirable to have a dispersion relation for surface plasmons which does not depend, in its derivation, on some particular assumption about the nature of the interaction of an electron with the boundary. Moreover, it would also seem to be desirable to have such a dispersion relation which is not based on the assumption that the dielectric properties of the medium supporting the surface plasmon are due to a specific collective excitation in the medium, viz. an electron plasma, so that it applies to systems in which these properties are associated with other electric dipole excitations such as IR active phonons and excitons, for example, or combinations of them.

It seemed to be worthwhile, therefore, to try to obtain a dispersion relation for surface plasmons in the presence of spatial dispersion of a form similar to that of Eq. (1.1), but without invoking the restrictive assumption of specular reflection of electrons at the boundary of the dielectric, in which the central role is played by the nonlocal dielectric constant of the medium, about which a minimum number of assumptions are made.

In this paper we present the derivation of such a dispersion relation. It is obtained in Section II, and its use illustrated by application to systems for which the dispersion relation is already known in Section III. A discussion of the results obtained is given in Section IV.

II. The Dispersion Relation

We assume a dielectric medium which occupies the semi-infinite region $x_3 > 0$. The region $x_3 < 0$ is occupied by vacuum. Because the system possesses infinitesimal translational invariance in directions parallel to the surface, the macroscopic electric field and the displacement in it can be expressed in the forms

$$\vec{E}(\vec{x},t) = \vec{E}(\vec{k}_\parallel\omega|x_3)e^{i\vec{k}_\parallel\cdot\vec{x}_\parallel - i\omega t} \tag{2.1a}$$

$$\vec{D}(\vec{x},t) = \vec{D}(\vec{k}_\parallel\omega|x_3)e^{i\vec{k}_\parallel\cdot\vec{x}_\parallel - i\omega t} \quad , \tag{2.1b}$$

where $\vec{k}_\parallel = \hat{x}_1k_1 + \hat{x}_2k_2$, $\vec{x}_\parallel = \hat{x}_1x_1 + \hat{x}_2x_2$, and $\hat{x}_1$ and $\hat{x}_2$ are two mutually perpendicular unit vectors in the plane of the dielectric-vacuum interface. Within the dielectric medium the relation between $\vec{D}(\vec{k}_\parallel\omega|x_3)$ and $\vec{E}(\vec{k}_\parallel\omega|x_3)$ is assumed to be

$$\vec{D}(\vec{k}_\parallel\omega|x_3) = \int_0^\infty dx_3'\epsilon(\vec{k}_\parallel\omega|x_3x_3')\vec{E}(\vec{k}_\parallel\omega|x_3') \qquad x_3 \geq 0 \quad , \tag{2.2}$$

where $\epsilon(\vec{k}_\parallel\omega|x_3x_3')$ is assumed to be symmetric in the variables x_3 and x_3'.

Thus, we assume the nonlocal dielectric tensor of the semi-infinite medium to be isotropic. This assumption is similar to and, because of the lower symmetry of the semi-infinite medium, may be as restrictive as the combined assumption by Ritchie and Marasuk of an isotropic, nonlocal, bulk dielectric tensor and specular reflection of electrons from the boundary. The assumption of isotropy, however, can be removed from both the present theory and that of Ritchie and Marusak. The result in each case is a more complicated dispersion relation. The assumption of specular reflection from the boundary, on the other hand, is essential in the Ritchie-Marusak approach, while it is not in the present treatment.

The Maxwell equation

$$\nabla \cdot \vec{D}(\vec{x},t) = 0 \tag{2.3}$$

is now combined with Eq. (2.2) and with the equation of the electrostatic approximation

$$\vec{E}(\vec{x},t) = -\nabla\varphi(\vec{x},t) \quad , \tag{2.4}$$

where $\varphi(\vec{x},t)$ is a scalar potential, to yield an integro-differential equation for $\varphi(\vec{k}_{\parallel}\omega|x_3)$, the Fourier coefficient of $\varphi(\vec{x},t)$:

$$k_{\parallel}^2 \int_0^{\infty} dx_3' \epsilon(\vec{k}_{\parallel}\omega|x_3x_3')\varphi(\vec{k}_{\parallel}\omega|x_3') - \frac{d}{dx_3}\int_0^{\infty} dx_3' \epsilon(\vec{k}_{\parallel}\omega|x_3x_3') \frac{d}{dx_3'}\varphi(\vec{k}_{\parallel}\omega|x_3') = 0 \; . \tag{2.5}$$

To solve Eq. (2.5) we begin by formally expanding both $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$ and $(d/dx_3)\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$ in double series of functions complete and orthonormal in the semi-infinite interval $(0,\infty)$, and localized in the vicinity of the interface at $x_3 = 0$:

$$\epsilon(\vec{k}_{\parallel}\omega|x_3x_3') = \sum_{m,n=0}^{\infty} a_{mn}(\vec{k}_{\parallel}\omega)\varphi_m(x_3)\varphi_n(x_3') \tag{2.6a}$$

$$\frac{d}{dx_3}\,\epsilon(\vec{k}_{\parallel}\omega|x_3x_3') = \sum_{m,n=0}^{\infty} b_{mn}(\vec{k}_{\parallel}\omega)\varphi_m(x_3)\varphi_n(x_3') \quad . \tag{2.6b}$$

Although our final result will be independent of any particular choice for the $\{\varphi_m(x_3)\}$, a convenient choice, for definiteness, is the set of Laguerre functions defined by

$$\varphi_n(x_3) = \beta^{\frac{1}{2}} e^{-\frac{1}{2}\beta x_3} \frac{L_n(\beta x_3)}{n!} \quad , \tag{2.7}$$

where $L_n(x_3)$ is the $n^{\underline{th}}$ Laguerre polynomial, and β is a real, positive parameter with the dimensions of an inverse length.

Because of our assumption that $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$ is symmetric in x_3 and x_3', it is the case that the coefficients $\{a_{mn}(k_{\parallel}\omega)\}$ are symmetric in the subscripts m and n.

We next expand $\varphi(\vec{k}_{\parallel}\omega|x_3)$ and $(d/dx_3)\varphi(\vec{k}_{\parallel}\omega|x_3)$ in terms of the $\{\varphi_m(x_3)\}$:

$$\varphi(\vec{k}_{\parallel}\omega|x_3) = \sum_{n=0}^{\infty} A_n(\vec{k}_{\parallel}\omega)\varphi_n(x_3) \tag{2.8a}$$

$$\frac{d}{dx_3}\,\varphi(\vec{k}_{\parallel}\omega|x_3) = \sum_{n=0}^{\infty} B_n(\vec{k}_{\parallel}\omega)\varphi_n(x_3) \quad , \tag{2.8b}$$

so that

$$A_n(\vec{k}_\parallel\omega) = \int_0^\infty dx_3 \varphi_n(x_3)\varphi(\vec{k}_\parallel\omega|x_3) \tag{2.9a}$$

$$B_n(\vec{k}_\parallel\omega) = \int_0^\infty dx_3 \varphi_n(x_3) \frac{d}{dx_3} \varphi(\vec{k}_\parallel\omega|x_3) \quad . \tag{2.9b}$$

When we substitute Eqs. (2.6) and (2.8) into Eq. (2.5) and use the orthonormality of the $\{\varphi_n(x_3)\}$, we find the latter takes the form

$$k_\parallel^2 \sum_{n=0}^\infty a_{mn}(\vec{k}_\parallel\omega)A_n(\vec{k}_\parallel\omega) = \sum_{n=0}^\infty b_{mn}(\vec{k}_\parallel\omega)B_n(\vec{k}_\parallel\omega) \quad . \tag{2.10}$$

The coefficients $\{A_n(\vec{k}_\parallel\varphi)\}$ and $\{B_n(\vec{k}_\parallel\omega)\}$ are not independent, however. If we integrate by parts the integral in Eq. (2.9b) we obtain the result that

$$B_n(\vec{k}_\parallel\omega) = -\varphi_n(0)\varphi(\vec{k}_\parallel\omega|0+) - \int_0^\infty dx_3\varphi(\vec{k}_\parallel\omega|x_3) \frac{d}{dx_3} \varphi_n(x_3) \quad , \tag{2.11}$$

where the notation $\varphi(\vec{k}_\parallel\omega|0+)$ is intended to emphasize that it is the potential on the medium side of the interface that is being considered. If we substitute Eq. (2.8a) into the second term on the right hand side of Eq. (2.11), we obtain finally the relation

$$B_n(\vec{k}_\parallel\omega) = -\varphi_n(0)\varphi(\vec{k}_\parallel\omega|0+) - \sum_{p=0}^\infty e_{np}A_p(\vec{k}_\parallel\omega) \quad , \tag{2.12}$$

where

$$e_{np} = \int_0^\infty dx_3 \left(\frac{d}{dx_3}\varphi_n(x_3)\right)\varphi_p(x_3) \quad . \tag{2.13a}$$

We note for future reference that e_{np} can be expressed equivalently as

$$e_{np} = \int_0^\infty dx_3 \int_0^\infty dx_3' \varphi_p(x_3) \frac{d}{dx_3} \delta(x_3 - x_3')\varphi_n(x_3') = - \varphi_p(0)\varphi_n(0) - e_{pn} \quad . \tag{2.13b}$$

On combining Eqs. (2.10) and (2.12) we find that the coefficient $B_n(\vec{k}_\parallel\omega)$ is given by

$$B_n(\vec{k}_\parallel\omega) = - k_\parallel^2\varphi(\vec{k}_\parallel\omega|0+) \sum_{p=0}^{\infty} \left[k_\parallel^2\overleftrightarrow{I} + \overleftrightarrow{e}\overleftrightarrow{a}^{-1}\overleftrightarrow{b}\right]^{-1}_{np}\varphi_p(0) \quad , \tag{2.14}$$

where for convenience we have suppressed the argument $(\vec{k}_\parallel\omega)$ of the matrices $\overleftrightarrow{a}(\vec{k}_\parallel\omega)$ and $\overleftrightarrow{b}(\vec{k}_\parallel\omega)$ on the right hand side of this equation. It follows from this result and Eqs. (2.2), (2.4), (2.6a) and (2.9b) that

$$D_3(\vec{k}_\parallel\omega|0+) = k_\parallel^2\varphi(\vec{k}_\parallel\omega|0+) \sum_{m,n=0}^{\infty} \varphi_m(0)\left[k_\parallel^2\overleftrightarrow{a}^{-1} + \overleftrightarrow{e}\overleftrightarrow{a}^{-1}\overleftrightarrow{b}\overleftrightarrow{a}^{-1}\right]^{-1}_{mn}\varphi_n(0) \quad . \tag{2.15}$$

We now turn to the vacuum region $x_3 < 0$. In this region the function $\varphi(\vec{k}_\parallel\omega|x_3)$ is readily found to have the form

$$\varphi(\vec{k}_\parallel\omega|x_3) = Ae^{k_\parallel x_3} \qquad x_3 < 0 \quad , \tag{2.16}$$

where A is an arbitrary constant, while the displacement component $D_3(\vec{k}_\parallel\omega|x_3)$ becomes

$$D_3(\vec{k}_\parallel\omega|x_3) = - Ak_\parallel e^{k_\parallel x_3} \qquad x_3 < 0 \quad . \tag{2.17}$$

The boundary conditions in the problem are the continuity of $\varphi(\vec{x},t)$ and $D_3(\vec{x},t)$ across the surface $x_3 = 0$. The first of these yields the relation $\varphi(\vec{k}_\parallel\omega|0+) = A$; the use of this result together with Eqs. (2.15) and (2.17) in the second boundary condition yields the dispersion relation for surface plasmons in the form

$$1 + k_\parallel \sum_{m,n=0}^{\infty} \varphi_m(0)\chi_{mn}(\vec{k}_\parallel\omega)\varphi_n(0) = 0 \quad , \tag{2.18a}$$

where

$$\chi_{mn}(\vec{k}_\parallel\omega) = \left[k_\parallel^2 \overleftrightarrow{a}^{-1} + \overleftrightarrow{ea}^{-1}\overleftrightarrow{ba}^{-1}\right]^{-1}_{mn} . \tag{2.18b}$$

It is useful to point out that $\chi_{mn}(\vec{k}_\parallel\omega)$ is symmetric in m and n. For this purpose it suffices to show that the matrix $k_\parallel^2\overleftrightarrow{a}^{-1} + \overleftrightarrow{ea}^{-1}\overleftrightarrow{ba}^{-1}$ is a symmetric matrix. Inasmuch as the matrix $\overleftrightarrow{a}^{-1}$ is symmetric because the matrix $\overleftrightarrow{a}$ is, it is necessary to show only that the matrix $\overleftrightarrow{X} = \overleftrightarrow{ea}^{-1}\overleftrightarrow{ba}^{-1}$ is symmetric.

To show this we first relate the coefficients $\left\{b_{mn}(\vec{k}_\parallel\omega)\right\}$ to the coefficients $\left\{a_{mn}(\vec{k}_\parallel\omega)\right\}$. We see from Eq. (2.6b) that

$$b_{mn}(\vec{k}_\parallel\omega) = \int_0^\infty dx_3 \int_0^\infty dx_3'\varphi_m(x_3)\ \frac{d}{dx_3}\ \epsilon(\vec{k}_\parallel\omega|x_3x_3')\varphi_n(x_3') \quad . \tag{2.19}$$

An integration by parts yields

$$b_{mn}(\vec{k}_\parallel\omega) = -\int_0^\infty dx_3'\varphi_m(o)\epsilon(\vec{k}_\parallel\omega|ox_3')\varphi_n(x_3') - \\ - \int_0^\infty dx_3 \int_0^\infty dx_3' \left(\frac{d}{dx_3}\ \varphi_m(x_3)\right)\epsilon(\vec{k}_\parallel\omega|x_3x_3')\varphi_n(x_3') \quad . \tag{2.20}$$

We now substitute Eq. (2.6a) into the right hand side of this equation, and use Eq. (2.13a) to obtain

$$b_{mn}(\vec{k}_{\parallel}\omega) = -\sum_{r=o}^{\infty} \left\{\varphi_m(o)\varphi_r(o) + e_{mr}\right\} a_{rn}(\vec{k}_{\parallel}\omega)$$

$$= \sum_{r} e_{rm}\, a_{rn}(\vec{k}_{\parallel}\omega) \quad . \tag{2.21}$$

The second equality in Eq. (2.21) follows from the use of Eq. (2.13b).

It follows from Eq. (2.21) that the matrix X can be written equivalently as

$$\overleftrightarrow{X} = \overleftrightarrow{e}\overleftrightarrow{a}^{-1}(\overleftrightarrow{e}^{T}\overleftrightarrow{a})\overleftrightarrow{a}^{-1} = \overleftrightarrow{e}\overleftrightarrow{a}^{-1}\overleftrightarrow{e}^{T} = \overleftrightarrow{X}^{T} , \tag{2.22}$$

where $\overleftrightarrow{M}^{T}$ denotes the transpose of the matrix $\overleftrightarrow{M}$. The matrix $\overleftrightarrow{\chi}(\vec{k}_{\parallel}\omega)$ is therefore symmetric, and can be written alternatively as

$$\chi_{mn}(\vec{k}_{\parallel}\omega) = \left[k_{\parallel}^{2}\overleftrightarrow{a}^{-1} + \overleftrightarrow{e}\overleftrightarrow{a}^{-1}\overleftrightarrow{e}^{T}\right]^{-1}_{mn} \quad . \tag{2.18c}$$

Equation (2.18) may have some interest of its own in connection with purely numerical studies of surface plasmons in spatially dispersive media. However, at this point we direct our efforts to re-expressing Eq. (2.18) in a form in which the $\left\{\varphi_m(x_3)\right\}$ no longer appear explicitly or implicitly.

We begin by introducing the function $\chi(\vec{k}_{\parallel}\omega|x_3x_3')$, which is defined by

$$\chi(\vec{x}_{\parallel}\omega|x_3x_3') = \sum_{m,n=o}^{\infty} \varphi_m(x_3)\chi_{mn}(\vec{k}_{\parallel}\omega)\varphi_n(x_3') \qquad x_3,x_3' > 0. \tag{2.23}$$

In terms of this function the dispersion relation (2.18) becomes

$$1 + k_{\parallel}\,\chi(\vec{k}_{\parallel}\omega|oo) = 0. \tag{2.24}$$

To obtain the function $\chi(\vec{k}_\parallel\omega|x_3x_3')$ or more accurately, the equations determining it, we return to Eq. (2.18c), which we rewrite as

$$k_\parallel^2 \sum_p a_{mp}^{-1}\chi_{pn} + \sum_{prs} e_{mp}a_{pr}^{-1}e_{sr}\chi_{sn} = \delta_{mn}. \tag{2.25}$$

We now multiply both sides of this equation by $\varphi_m(x_3)\varphi_n(x_3')$ and sum on m and n from zero to infinity. We recall the orthonormality and completeness conditions

$$\delta_{mn} = \int_o^\infty dx_3\varphi_m(x_3)\varphi_n(x_3) \tag{2.26a}$$

$$\sum_{m=o}^{\infty}\varphi_m(x_3)\varphi_m(x_3') = \delta(x_3 - x_3'). \tag{2.26b}$$

We next note that

$$\sum_{m,n=o}^{\infty}\varphi_m(x_3)a_{mn}^{-1}(\vec{k}_\parallel\omega)\varphi_n(x_3') = \epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3'), \tag{2.27}$$

where $\epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3')$ is the inverse of $\epsilon(\vec{k}_\parallel\omega|x_3x_3')$ in the sense that

$$\int_o^\infty dx_3''\ \epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3'')\epsilon\ (\vec{k}_\parallel\omega|x_3''x_3')$$

$$= \int_o^\infty dx_3''\epsilon\ (\vec{k}_\parallel\omega|x_3x_3'')\epsilon^{-1}(\vec{k}_\parallel\omega|x_3''x_3') = \delta(x_3-x_3'). \tag{2.28}$$

We also use the relations

$$\sum_{m,n=o}^{\infty}\varphi_m(x_3)e_{mn}\varphi_n(x_3') = -\left[\frac{d}{dx_3}\delta(x_3-x_3') + \delta(x_3)\delta(x_3')\right] \tag{2.29a}$$

$$\sum_{m,n=o}^{\infty}\varphi_m(x_3)e_{nm}\varphi_n(x_3') = \frac{d}{dx_3}\delta(x_3 - x_3'). \tag{2.29b}$$

The result of all of this is to transform Eq. (2.25) into

$$k_\parallel^2 \int_0^\infty dx_3'' \epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3'')\chi(\vec{k}_\parallel\omega|x_3''x_3') -$$

$$- \frac{d}{dx_3}\int_0^\infty dx_3''\ \epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3'')\frac{d}{dx_3''}\ \chi(\vec{k}_\parallel\omega|x_3''x_3') -$$

$$- \delta(x_3)\int_0^\infty dx_3''\ \epsilon^{-1}(\vec{k}_\parallel\omega|0x_3'')\frac{d}{dx_3''}\ \chi(\vec{k}_\parallel\omega|x_3''x_3') = \delta(x_3-x_3'). \qquad (2.30)$$

Finally, we see that the function $\chi(\vec{k}_\parallel\omega|x_3x_3')$ which satisfies the integro-differential equation

$$k_\parallel^2 \int_0^\infty dx_3''\ \epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3'')\chi(\vec{k}_\parallel\omega|x_3''x_3') -$$

$$- \frac{d}{dx_3}\int_0^\infty dx_3''\ \epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3'')\ \frac{d}{dx_3''}\chi(\vec{k}_\parallel\omega|x_3''x_3') = \delta(x_3-x_3'), \qquad (2.31a)$$

together with the boundary condition

$$\int_0^\infty dx_3''\ \epsilon^{-1}(\vec{k}_\parallel\omega|0\ x_3'')\ \frac{d}{dx_3''}\ \chi(\vec{k}_\parallel\omega|x_3''x_3') = 0, \qquad (2.31b)$$

is a solution of Eq. (2.30). As we are seeking electric fields localized in the vicinity of the surface $x_3 = 0$, the boundary condition which must be imposed on $\chi(\vec{k}_\parallel\omega|x_3x_3')$ at infinity is that

$$\lim_{x_3\to\infty} \chi(\vec{k}_\parallel\omega|x_3x_3') = 0. \qquad (2.31c)$$

Equation (2.24), together with the prescription for obtaining $\chi(\vec{k}_\parallel\omega|x_3x_3')$ given by Eqs. (2.31), is the central result of this section: the surface plasmon dispersion relation. In the next section we illustrate its use by applying it to two cases for which the surface plasmon dispersion relation is known.

III. Examples

In this section we solve Eqs. (2.31) for two choices of $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$, one local and the other nonlocal, for which the surface plasmon dispersion relation is already known, to illustrate the use of the dispersion relation, Eq. (2.24).

A. A Local Dielectric Tensor

We first consider the case in which $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$ is given by

$$\epsilon(\vec{k}_{\parallel}\omega|x_3x_3') = \epsilon(\omega)\delta(x_3 - x_3'), \tag{3.1}$$

where $\epsilon(\omega)$ is independent of $\vec{k}_{\parallel}$ and of the coordinates x_3 and x_3'. It follows that

$$\epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x_3') = \frac{1}{\epsilon(\omega)}\,\delta(x_3 - x_3'). \tag{3.2}$$

The equation satisfied by $\chi(\vec{k}_{\parallel}\omega|x_3x_3')$ in this case becomes

$$\frac{k_{\parallel}^2}{\epsilon(\omega)}\,\chi(\vec{k}_{\parallel}\omega|x_3x_3') - \frac{1}{\epsilon(\omega)}\,\frac{d^2}{dx_3^2}\,\chi(\vec{k}_{\parallel}\omega|x_3x_3') = \delta(x_3 - x_3') \tag{3.3a}$$

together with the boundary condition

$$\frac{d}{dx_3}\,\chi(\vec{k}_{\parallel}\omega|x_3x_3')\Big|_{x_3=0} = 0. \tag{3.3b}$$

The solution of Eq. (3.3a) which vanishes as $x_3 \to \infty$ is

$$\chi(\vec{k}_{\parallel}\omega|x_3x_3') = -\epsilon(\omega)\left\{\frac{e^{-k_{\parallel}|x_3-x_3'|}}{-2k_{\parallel}} + Ae^{-k_{\parallel}x_3}\right\}, \tag{3.4}$$

where the first term is the particular integral and the second is the complementary function. The coefficient A is determined with the aid of Eq. (3.3b), with the result that

$$\chi(\vec{k}_{\parallel}\omega|x_3x_3') = \frac{\epsilon(\omega)}{2k_{\parallel}}\left\{e^{-k_{\parallel}|x_3-x_3'|} + e^{-k_{\parallel}(x_3+x_3')}\right\}. \tag{3.5}$$

When Eq. (3.5) is substituted into Eq. (2.24), we obtain as the surface plasmon dispersion relation

$$1 + \epsilon(\omega) = 0, \tag{3.6}$$

a well-known result for this case.[3]

B. A Nonlocal Dielectric Constant

The second example we consider is based on the nonlocal dielectric function

$$\epsilon(\vec{k}_{\parallel}\omega|x_3x_3') = \epsilon_o\delta(x_3-x_3') + i\,\frac{\omega_p^2}{2D\Gamma}\,e^{i\Gamma|x_3-x_3'|} \qquad x_3,x_3' \geq 0\ , \tag{3.7}$$

which was used by Birman and Sein[4], Maradudin and Mills[5], and Agarwal, Pattanayak, and Wolf[6], in studies of various optical optical properties of semi-infinite media in the presence of spatial dispersion. There are some unphysical features of the model underlying this form for $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$, as will be discussed in the following section. However, because results based on its use are available it serves as a useful example to illustrate Eq. (2.24), which is not restricted by these unphysical features. In Eq. (3.7) ϵ_o is the optical frequency dielectric constant, which is assumed to be real and frequency independent; ω_p is a plasma frequency; D is a positive constant which defines the curvature of the exciton or transverse optical phonon dispersion curve at $\vec{k} = o$; and Γ is given by

$$\Gamma = \left(\frac{\omega^2-\omega_o^2}{D} - k_\parallel^2 + \frac{i\omega\gamma}{D}\right)^{\frac{1}{2}} , \tag{3.8}$$

so that Re $\Gamma > 0$, Im $\Gamma > 0$. The frequency ω_o in Eq. (3.8) is that of the exciton or transverse optical phonon at $\vec{k} = 0$.

The equation for the inverse dielectric function $\epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3')$, Eq. (2.28), in the present case takes the form

$$\epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3') + \frac{i\omega_p^2}{2\epsilon_o D\Gamma}\int_o^\infty dx_3'' \, e^{i\Gamma|x_3-x_3''|}\epsilon^{-1}(\vec{k}_\parallel\omega|x_3''x_3') = \epsilon_o^{-1}\delta(x_3-x_3'). \tag{3.9}$$

We apply the operator $(d^2/dx_3^2) + \Gamma^2$ to both sides of this equation to transform it into

$$\left(\frac{d^2}{dx_3^2} + \alpha^2\right)\epsilon^{-1}(\vec{k}_\parallel\omega|x_3x_3') = \frac{1}{\epsilon_o}\left(\frac{d^2}{dx_3^2} + \Gamma^2\right)\delta(x_3-x_3'), \tag{3.10}$$

where

$$\alpha = \left(\frac{\omega^2-\omega_L^2}{D} - k_\parallel^2 + \frac{i\omega\gamma}{D}\right)^{\frac{1}{2}} \tag{3.11a}$$

and

$$\omega_L^2 = \omega_o^2 + (\omega_p^2/\epsilon_o), \tag{3.11b}$$

so that Re $\alpha > 0$, Im $\alpha > 0$. We now use the fact that

$$\left(\frac{d^2}{dx_3^2} + \alpha^2\right)\left\{\delta(x_3 - x_3') + \frac{\Gamma^2-\alpha^2}{2i\alpha}\, e^{i\alpha|x_3-x_3'|}\right\} = \left(\frac{d^2}{dx_3^2} + \Gamma^2\right)\delta(x_3 - x_3') \tag{3.12}$$

to write the solution of Eq. (3.10) in the form

$$\epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x_3') = \frac{1}{\epsilon_o}\,\delta(x_3-x_3') + \frac{\omega_p^2}{\epsilon_o^2 D}\,\frac{e^{i\alpha|x_3-x_3'|}}{2i\alpha} + Ae^{i\alpha x_3}. \tag{3.13}$$

The coefficient A is obtained by requiring that the expression (3.13) also satisfy the original integral equation (3.9). In this way we obtain finally the result that

$$\begin{aligned}\epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x_3') &= \frac{1}{\epsilon_o}\,\delta(x_3-x_3') + \\ &+ \frac{\omega_p^2}{\epsilon_o^2 D}\,\frac{1}{2i\alpha}\left[e^{i\alpha|x_3-x_3'|} + \frac{\alpha-\Gamma}{\alpha+\Gamma}\,e^{i\alpha(x_3+x_3')}\right] \\ &\equiv \frac{1}{\epsilon_o}\,\delta(x_3-x_3') + f(\vec{k}_{\parallel}\omega|x_3x_3').\end{aligned} \tag{3.14}$$

We note the result that

$$\left(\frac{d^2}{dx_3^2} + \alpha^2\right)f(\vec{k}_{\parallel}\omega|x_3x_3') = \frac{\omega_p^2}{\epsilon_o^2 D}\,\delta(x_3-x_3'). \tag{3.15}$$

The integral equation (2.31a) for the function $\chi(\vec{k}_{\parallel}\omega|x_3x_3')$ now takes the form

$$\begin{aligned}&\frac{1}{\epsilon_o}\left(\frac{d^2}{dx_3^2} - k_{\parallel}^2\right)\chi(\vec{k}_{\parallel}\omega|x_3x_3') + \frac{d}{dx_3}\int_0^\infty dx_3'' f(\vec{k}_{\parallel}\omega|x_3x_3'')\,\times \\ &\times\frac{d}{dx_3''}\,\chi(\vec{k}_{\parallel}\omega|x_3''x_3') - k_{\parallel}^2\int_0^\infty dx_3'' f(\vec{k}_{\parallel}\omega|x_3x_3'')\chi(\vec{k}_{\parallel}\omega|x_3''x_3') = \\ &= -\delta(x_3-x_3').\end{aligned} \tag{3.16}$$

We now apply the operator $(d^2/dx_3^2) + \alpha^2$ to both sides of this equation, and use Eq. (3.15), to obtain

$$\left(\frac{d^2}{dx_3^2} - k_\parallel^2\right)\left(\frac{d^2}{dx_3^2} - \beta^2\right)\chi(\vec{k}_\parallel\omega|x_3x_3') = -\epsilon_o\left(\frac{d^2}{dx_3^2} + \alpha^2\right)\delta(x_3-x_3'), \tag{3.17}$$

where

$$\beta = \left(\frac{\omega_o^2-\omega^2}{D} + k_\parallel^2 - \frac{i\omega\gamma}{D}\right)^{\frac{1}{2}} = -i\Gamma, \tag{3.18}$$

so that Re $\beta > 0$, Im $\beta < 0$. The solution of Eq. (3.17) which vanishes at infinity is given by

$$\chi(\vec{k}_\parallel\omega|x_3x_3') = -\frac{\epsilon_o}{\beta^2 - k_\parallel^2}\left[(\alpha^2+k_\parallel^2)\,\frac{e^{-k_\parallel|x_3-x_3'|}}{2k_\parallel} - \right.$$

$$\left. - (\alpha^2+\beta^2)\,\frac{e^{-\beta|x_3-x_3'|}}{2\beta}\right] + C_1e^{-k_\parallel x_3} + C_2e^{-\beta x_3}. \tag{3.19}$$

The two coefficients C_1 and C_2 in this solution are determined from the two equations obtained by requiring that the solution (3.19) satisfy the integro-differential equation (3.16) on the one hand, and from the boundary condition (2.31b) on the other. After some tedious analysis the following result is obtained:

$$\chi(\vec{k}_\parallel\omega|x_3x_3') = \frac{\epsilon(\omega)}{2k_\parallel}\left[e^{-k_\parallel|x_3-x_3'|} + \right.$$

$$\left. + \frac{ik_\parallel+\alpha}{ik_\parallel-\alpha}\,\frac{ik_\parallel-\Gamma}{ik_\parallel+\Gamma}\,e^{-k_\parallel(x_3+x_3')}\right] + \frac{(\epsilon(\omega) - \epsilon_o)}{2i\Gamma}\,e^{i\Gamma|x_3-x_3'|}, \tag{3.20}$$

where

$$\epsilon(\omega) = \epsilon_o + \frac{\omega_p^2}{\omega_o^2 - \omega^2 - i\omega\gamma} \tag{3.21}$$

is the bulk dielectric constant at $\vec{k} = 0$, and where Eq. (3.18) has been used.

Substitution of Eq. (3.20) into Eq. (2.24) yields as the dispersion relation for surface plasmons

$$1 + \frac{\epsilon(\omega)}{2}\left[1 + \frac{ik_\parallel+\alpha}{ik_\parallel-\alpha}\,\frac{ik_\parallel-\Gamma}{ik_\parallel+\Gamma}\right] + \frac{k_\parallel}{2i\Gamma}(\epsilon(\omega) - \epsilon_o) = 0. \tag{3.22}$$

This equation can be rearranged into the form

$$1 + \epsilon(\omega) = ik_\parallel\left[\epsilon(\omega)\,\frac{\alpha - \Gamma}{(k_\parallel+i\alpha)(k_\parallel-i\Gamma)} + \frac{\epsilon(\omega)-\epsilon_o}{2\Gamma}\right] . \tag{3.23}$$

Inasmuch as the right hand side of this equation is explicitly proportional to $k_\parallel$, in obtaining the frequency of the surface plasmon to lowest nonvanishing order in $k_\parallel$ it suffices to set $k_\parallel = 0$ in the expression in braces. We thus obtain the approximate dispersion relation

$$1 + \epsilon(\omega) \cong ik_\parallel\left[\epsilon(\omega)\left(\frac{1}{\Gamma} - \frac{1}{\alpha}\right) + \frac{\epsilon(\omega) - \epsilon_o}{2\Gamma}\right]_{k_\parallel=0} . \tag{3.24}$$

At this point we neglect the intrinsic damping in the dielectric medium and let γ tend to zero through positive values. In this limit we find that

$$\begin{aligned} \Gamma(k_\parallel = 0) &= iD^{-\frac{1}{2}}(\omega_o^2 - \omega^2)^{\frac{1}{2}} && \omega < \omega_o \\ &= D^{-\frac{1}{2}}(\omega^2 - \omega_o^2) && \omega > \omega_o \end{aligned} \tag{3.25}$$

$$\begin{aligned} \alpha(k_\parallel = 0) &= iD^{-\frac{1}{2}}(\omega_L^2 - \omega^2)^{\frac{1}{2}} && \omega < \omega_L \\ &= D^{-\frac{1}{2}}(\omega^2 - \omega_L^2)^{\frac{1}{2}} && \omega > \omega_L \end{aligned} \tag{3.26}$$

We next note that the equation $\epsilon(\omega) + 1 = 0$ is the surface plasmon dispersion relation in the absence of spatial dispersion. Consequently the frequency of the surface plasmon in the absence of spatial dispersion, ω_s, is the solution of the equation

$$\epsilon(\omega_s) + 1 = 0 \quad . \tag{3.27}$$

We thus write the solution of Eq. (3.24) in the form

$$\omega = \omega_s + \Delta\omega_s \, , \tag{3.28}$$

where $\Delta\omega_s$ is of first order in $k_\parallel$. Substituting Eq. (3.28) into Eq. (3.24) and linearizing the resulting equation in $\Delta\omega_s$ with the aid of Eq. (3.27) we obtain

$$\Delta\omega_s \cong \frac{ik_\parallel}{\epsilon'(\omega_s)} \left[\frac{1}{\alpha} - \frac{3+\epsilon_o}{2\Gamma} \right]_{\substack{k_\parallel = 0 \\ \omega = \omega_s}} \tag{3.29}$$

If we denote the static ($\omega = 0$) value of the bulk dielectric constant at $\vec{k} = 0$ by ϵ_s, we have the relation

$$\epsilon_s = \epsilon_o + (\omega_p^2/\omega_o^2) \quad . \tag{3.30}$$

With the substitution of Eq. (3.21) (with $\gamma = 0$) into Eq. (3.27) we find that the frequency of the surface plasmon in the absence of spatial dispersion is given by

$$\omega_s^2 = \omega_o^2 + \frac{\omega_p^2}{1+\epsilon_o} \, , \tag{3.31a}$$

or equivalently

$$\omega_s = \left[\frac{1+\epsilon_s}{1+\epsilon_o}\right]^{\frac{1}{2}} \omega_o \quad . \tag{3.31b}$$

It is readily determined that

$$\omega_o < \omega_s < \omega_L \quad . \tag{3.32}$$

These inequalities, together with Eqs. (3.25) and (3.26), enable us to express $\Delta\omega_s$ given by Eq. (3.29) in the form

$$\begin{aligned} \Delta\omega_s &= \frac{D^{\frac{1}{2}}k_\parallel}{\epsilon'(\omega_s)}\left[\frac{1}{(\omega_L^2-\omega_s^2)^{\frac{1}{2}}} - \frac{i(\epsilon_o+3)}{2}\frac{1}{(\omega_s^2-\omega_o^2)^{\frac{1}{2}}}\right] \\ &= \frac{D^{\frac{1}{2}}(1+\epsilon_o)^{\frac{1}{2}}k_\parallel}{\omega_p\epsilon'(\omega_s)}\left[\epsilon_o^{\frac{1}{2}} - i\,\frac{\epsilon_o+3}{2}\right] \quad , \end{aligned} \tag{3.33}$$

which agrees completely with the result for $\Delta\omega_s$ obtained by Maradudin and Mills[5] on letting the speed of light tend to infinity in the surface polariton dispersion relation they obtained on the basis of the dielectric constant (3.7).

IV. Discussion

In this paper we have obtained an explicit dispersion relation for surface plasmons at the interface between vacuum and a semi-infinite dielectric medium occupying the half-space $x_3 > 0$ and characterized by a nonlocal dielectric constant $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$. It should be remarked that the derivation of Eqs. (2.24) and (2.31) presented here is somewhat artificial. It should be possible to obtain these results more directly, but we have not attempted to do so. It also appears as if the use of Eqs. (2.24) and (2.31) to obtain the surface plasmon dispersion relation may involve lengthier calculations than are required in a more conventional approach based on a direct solution of the equations of electrostatics. Nevertheless, it still seems useful to have an explicit dispersion relation, Eq. (2.24), with a definite prescription for obtaining the function $\chi(\vec{k}_{\parallel}\omega|x_3x_3')$ entering it, Eq. (2.31). For example, in the case of dielectric constants of more complicated form than those considered in the preceding section Eq. (2.31) can serve as the basis for approximate determinations of $\chi(\vec{k}_{\parallel}\omega|x_3x_3')$, e.g. by variational methods.

The fact that the method developed here yields the same surface plasmon frequency, Eqs. (3.28), (3.31), and (3.33), as was obtained by a rather different method by Maradudin and Mills[5] is of some independent interest for the following reason. The dielectric constant (3.7) is obtained by partially Fourier transforming the bulk dielectric constant

$$\epsilon(\vec{k},\omega) = \epsilon_0 + \frac{\omega_p^2}{\omega_0^2 + Dk^2 - \omega^2 - i\omega\gamma} \qquad (4.1)$$

according to

$$\epsilon(\vec{k}_{\parallel}\omega|x_3x_3') = \int_{-\infty}^{\infty} \frac{dk_3}{2\pi}\, \epsilon(\vec{k},\omega)\, e^{ik_3(x_3 - x_3')} , \tag{4.2}$$

and restricting x_3 and x_3' to be positive. Recently Horing[7] used $\epsilon(\vec{k},\omega)$ as given by Eq. (4.1) in the Ritchie-Marusak dispersion relation, Eq. (1.1), and obtained an expression for the frequency of a surface plasmon which differs from the Maradudin-Mills result, Eqs. (3.28), (3.31), and (3.33). This difference is not very surprising in fact. As we have noted in the Introduction, underlying the Ritchie-Marusak dispersion relation is the assumption that electrons are reflected specularly from the surface of the solid. The dielectric constant (3.7), on the other hand, represents the so-called dielectric approximation, in which the bulk dielectric constant is partially Fourier transformed according to Eq. (4.2), after which x_3 and x_3' are restricted to be positive, so that the resulting $\epsilon(\vec{k}_{\parallel}\omega|x_3x_3')$ is no longer a function of $x_3 - x_3'$, but depends on x_3 and x_3' separately. It is known that the dielectric approximation does not conserve particle number[8], and causes the surface to act as a source or [illegible] of energy[9], neither of which is the case when specular [illegible] is assumed. Thus a different physical situation is being considered in Horing's work from that assumed in the work of Maradudin and Mills. Consequently, it is not surprising that different surface plasmon frequencies are obtained in these two calculations. However, this example points up the desirability of having a dispersion relation for surface plasmons which is valid for more general physical situations than is represented by the assumption of specular reflection at the surface. Equations (2.24) and (2.31) provide such a dispersion relation.

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